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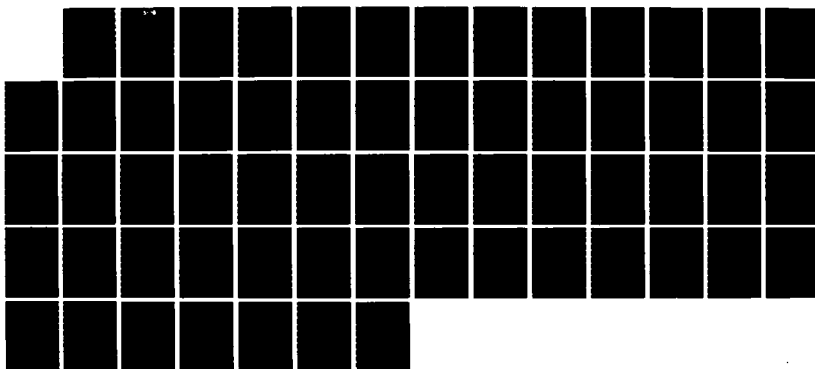
PICOSECOND NONLINEAR RESONANT INTERACTIONS IN
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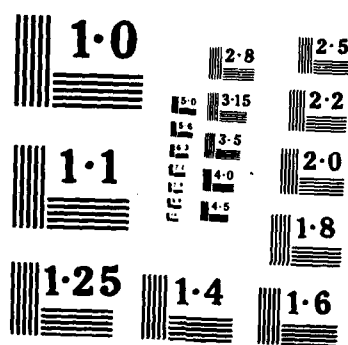
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<p> This research was aimed at advancing understanding and utilization of selected optical properties of semiconductors containing magnetic elements. Emphasis was placed on the interaction of such materials with ultrashort pulses of laser radiation in order to study coupled electronic and magnetic excitations under selected nonequilibrium conditions. We hoped to generate novel results through experimental research for applications to fast optoelectronic devices. The mixed crystal semiconductors (Cd, Mn)Se and (Cd, Mn)Te were used. The contract work has generated a number of "firsts", e.g. we measured the formation of local, microscopic magnetically oriented "domains" through real-time spectroscopy with picosecond laser pulses. </p>			
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PICOSECOND NONLINEAR RESONANT INTERACTIONS IN SEMICONDUCTORS


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laboratories. In later phases of the contract research we have investigated the optical properties of a new semiconductor material, (Pb,Eu)Te, which has been shown to be of significance in infrared light emitting applications. In ultrathin layers of this material, we have also characterized the electronic properties in the so-called 'super-lattice structures'.

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1. Research Objectives and Summary

The objective of the research under AFOSR Contract F49620-82 C-0044 (01/01/82 to 03/31/86) was aimed at advancing the understanding and utilization of selected optical properties of semiconductors containing magnetic elements. In particular, emphasis was placed on the interaction of such materials with ultrashort pulses of laser radiation in order to study coupled electronic and magnetic excitations under selected nonequilibrium conditions. We hoped to be able to generate novel results through experimental research which might have applications for future fast optoelectronic devices. The mixed crystal semiconductors (Cd,Mn)Se and (Cd,Mn)Te have provided the material basis for our work. The contract work has been quite successful in generating a number of 'firsts' during the investigations of magneto-optical properties of these and related semiconductors by picosecond laser pulses. For example, we have measured the formation of local, microscopic magnetically oriented 'domains', the so-called bound magnetic polaron effects, through real-time spectroscopy. This has been an accomplishment very much the first of its kind and has triggered follow up work in several other leading laboratories. In later phases of the contract research we have investigated the optical properties of a new semiconductor material, (Pb,Eu)Te, which has been shown to be of significance in infrared light emitting applications. In ultrathin layers of this material, we have also characterized the electronic properties in the so-called 'superlattice structures'.

The research results derived from this AFOSR sponsored research have formed the basis of a number of scientific publications, as enumerated below.

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below. In addition to regular scientific meetings, the principal investigator has been invited to present the research results in several internationally recognized forums.

2. Research Accomplishments and Results

(a) The Exciton-Magnetic Polaron Effect in (Cd,Mn)Se

In this phase of the AFOSR supported research we have focussed on one of the more interesting effects encountered in the II-VI 'semimagnetic' semiconductors, namely the possibility of a finite magneto-optical contribution even at zero external magnetic field. Influence by the Mn⁺⁺-ion 3d electrons via an effective field on shallow impurities has been observed for donors in CdMnSe in spin-flip Raman scattering (1) and far-infrared spectroscopy (2), and for acceptors in CdMnTe in photoluminescence (3), (4). Theoretically, the inclusion of the exchange interaction of the impurity carrier with the Mn-ion spins within its orbit has been shown to lead to energy shifts and spin-splitting of the one-electron optical transition probability involving the impurity (5),(6),(1). These can originate from a net nonzero spin magnetization due to a trend toward local magnetic ordering within the Bohr volume of the carrier ('bound magnetic polaron') or a finite volume size effect which leaves a finite instantaneous magnetic moment even when thermal disorder dominates.

In the case of the wurtzite n-CdMnSe, recent work of Heiman, Wolff, and Warnock has clearly shown how the relatively large size of the donor orbit (>50Å) leads to a dominance of its zero field optical properties by the thermodynamic fluctuations even at a low temperature (6). For a photoluminescence event involving an exciton bound to the impurity, however, a question arises about the modification of the total exchange

interaction by the additional electron-hole pair. This is decidedly a difficult theoretical problem, but there is some experimental evidence to suggest that such effects may indeed be significant (7), (6). We have examined photoluminescence associated with the donor bound exciton in n-CdMnSe ($x=.10$ and $x=.05$) at zero external magnetic field and compared it with the same excitation in CdSe. In particular, we find that the presence of the Mn⁺⁺-ions affects a contribution to the binding energy and the linewidth of the bound exciton which is significantly larger than those observed for the 'bare' donor case (6). This is proposed to occur mainly as a consequence of a fairly temperature insensitive effective internal field within the relatively small exciton volume. We also find that the bound exciton luminescence spectrum for the $x=.10$ concentration shows a dependence on temperature which appears to be associated with a phase transition from an induced bound magnetic polaron regime. In our interpretation this is accompanied by large, inhomogeneously broadened spin-splittings which are reflected in anomalous spectral lineshapes and widths.

The experimental arrangement employed a cw frequency doubled Nd:YAG as a source of excitation at $h\nu=2.34$ eV. Photoluminescence spectra were obtained in a conventional way with samples mounted in a liquid helium immersion dewar. Figure 1 shows the dominant portion of the spectra in the near bandedge region, obtained for $x=.10$ samples at different temperatures. Somewhat unusually, however, strong reshaping and line narrowing occurs in the temperature range of 1.8 to 10 K. For comparison spectra for $x=.05$ and pure CdSe are shown in the inset. Care was exercised to provide a sufficiently low excitation (1mW) to avoid intensity dependent effects otherwise clearly evident. Additional spectral features

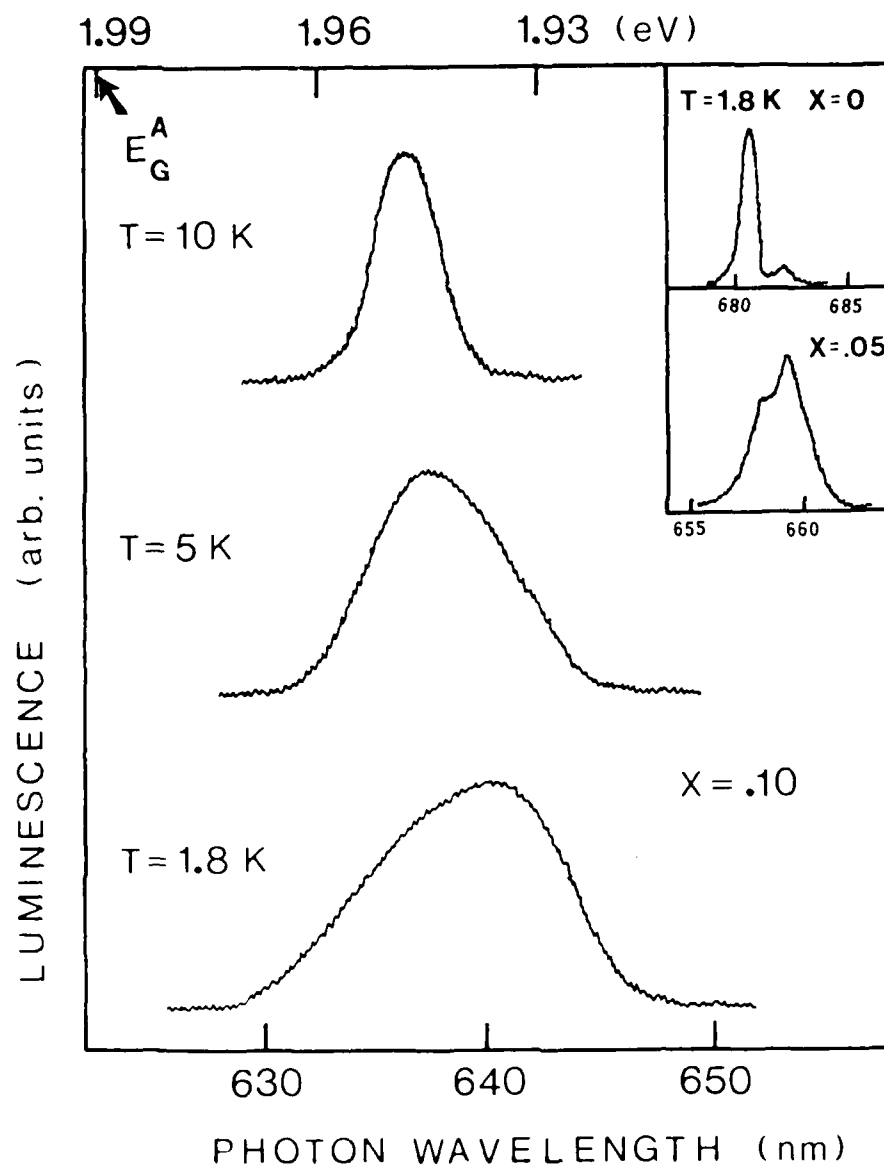


Figure 1: Low temperature luminescence spectra of (Cd,Mn)Se with $x = 0.10$; E_G^A indicates the bandgap energy for this composition. In the inset, spectra taken for $x = 0$ and $x = 0.05$ at the lowest temperature are shown for comparison.

were observable at lower photon energies, attributed to donor-acceptor pair transitions. The spectral component shown in Figure 1 for $x=.10$ is identified by us as the zero phonon dominated line of a neutral donor bound exciton from the A-valence band for the samples containing approximately 3×10^{16} donors per cm^{-3} . It shows a clear connection with the similar dominant feature in the $x=.05$ and $x=0$ samples. In comparison with the $x=.10$ material, the feature in the $x=.05$ samples exhibited a more clearly evident two peak substructure where the relative amplitudes varied with temperature in a manner expected for thermal redistribution. Figure 2 summarizes the energetic position of the luminescence maxima as a function of temperature for the three compositions, referenced to the corresponding bandgap energy. Apart from lineshape asymmetries, the graphed quantity E_p is a measure of the average binding energy of the bound exciton in each case. As shown elsewhere, temperature variation of the bandgap and the free exciton energy in the range of interest here is rather small (8),(9). At higher temperatures ($T \geq 50\text{K}$) the bound exciton feature in CdMnSe showed increased broadening in a manner expected from acoustic phonon interactions. Throughout the low temperature range all the spectra remained fairly strongly linearly polarized ($E \parallel c$), in a manner seen for the A-exciton in CdSe.

These experimental observations show that the donor bound exciton luminescence in our CdMnSe samples implies a significant increase in both the binding energy of the exciton and its linewidth, when compared with CdSe. Furthermore, the $x=.10$ material shows anomalous temperature behavior where the lineshape strongly alters and narrows from approximately 2 to 10 K. Insofar as the spin-splitting and the linewidth of the neutral donor for CdMnSe ($x \leq .10$) in the same samples was measured to be only about 1

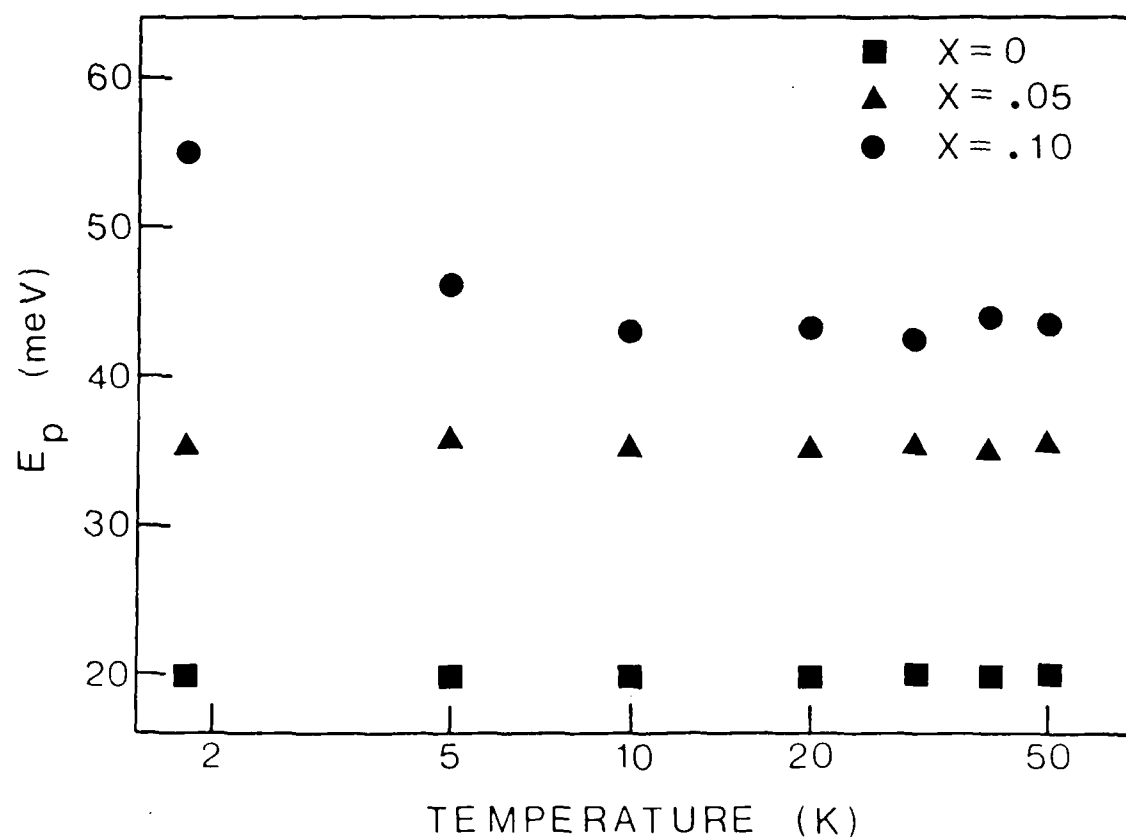


Figure 2: Energy position of the luminescence peaks as a function of temperature for three different compositions of (Cd,Mn)Se samples, reference to the corresponding bandgap energy in each case.

meV by spin-flip Raman scattering (6), we conclude that the initial exciton state is the dominant source of broadening in the luminescence event. It is well known that in nonmagnetic semiconductor alloys bound exciton lines acquire additional width from alloy potential (compositional) fluctuations. This follows from the relatively small effective radii of bound excitons. While such effects are likely to influence the bound exciton in CdMnSe as well, we believe that the major part of the observed energy shifts and broadening originates from the exchange interactions in the exciton-Mn⁺⁺ ion system. Analogous to the single particle impurity model which has wide experimental and theoretical support, both a bound magnetic polaron and a finite size magnetization effect need be considered within an effective Bohr radius of the exciton about the donor. In each case a substantial internal 'effective' magnetic field gives here rise to a spin splitting of the exciton. Alloy compositional fluctuations increase in importance for smaller radii, and lead to strong inhomogeneous broadening. Recent susceptibility measurements for CdMnSe indicate that for the compositional range considered here the magnetization remains paramagnetic at all temperatures (7),(8). The details of an inhomogeneous lineshape would, however, be strongly influenced by those remanent antiferromagnetic Mn-Mn interactions associated with elementary Mn-cluster formation.

These considerations lead us to suggest that for the $x=.10$ samples the initial strong temperature behavior indicates a phase transition from an exciton induced bound magnetic polaron regime to a fluctuation dominated one. The large linewidth (>30 meV) observed at 2K reflects the importance of alloy compositional fluctuations in the large internal effective field. In contrast, the $x=.05$ material remains in the fluctuation dominated

regime even at the lowest temperatures. In this regime, relatively temperature independent binding energy and linewidth are anticipated. Results similar to ours in magnitude have been recently reported by Nhung and Planel for a one-electron optical transition involving the neutral acceptor in CdMnTe (4). The expected spin-splitting into two components can be clearly seen in the $x=.05$ material while being somewhat obscured by a larger inhomogeneous broadening in the $x=.10$ samples. The origin of the broadening is due to the compositional fluctuations in the alloy and are therefore larger for smaller effective exciton radii. The inhomogeneous character of the lineshape has been also recently verified by studying energy diffusion in direct time-resolved experiments employing picosecond techniques (9).

An elementary estimate can be made about the expected size of the linewidth of the bound exciton in the fluctuation dominated regime, in the hydrogenic effective mass model. A sizable spin splitting with a broad linewidth is expected for an exciton radius of approximately 20 Å and a semiquantitative agreement is obtained with the measurements. Such a zeroth order estimate provides a less satisfactory answer, however, with the transition to the bound magnetic polaron regime for the $x=.10$ material, evidently reflecting the fundamental complication associated with the multiparticle character of the exciton. The observation of polarized luminescence in accordance with selection rules for the A-exciton in CdSe implies that either these selection rules remain intact even in the rather high internal fields encountered (>100 kG) or there may be an 'easy' axis of magnetization for spontaneous alignment of the net local moments.

In summary, photoluminescence spectra associated with the neutral donor bound exciton in CdMnSe shows a significant enhancement in its binding energy and linewidth in comparison with the previously studied bare donor. This is attributed to increase in the exchange interaction with the Mn^{++} -ion spins within the exciton orbit. Apart from a general trend, significant questions remain concerning the details of the electron-hole configuration in the magnetic polaron regime.

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(b) Formation of the Bound Magnetic Polaron in (Cd,Mn)Se

In this phase of the AFOSR supported research we have obtained results where for the first time evidence for the formation of a bound magnetic polaron (BMP) is directly obtained in time-resolved experiments by picosecond laser spectroscopy. In particular, we have examined the time evolution of spectra associated with the neutral donor bound exciton in $n\text{-Cd}_{0.90}\text{Mn}_{0.10}\text{Se}$. Our recent results from cw photoluminescence work in $\text{Cd}_{1-x}\text{Mn}_x\text{Se}$ have shown how the BMP contribution to the energy of a neutral donor bound exciton is likely to be significantly larger than that for the neutral donor.¹ This is attributed to the smaller effective Bohr radius of the exciton and the larger exchange constant for the (outer valence) hole, and manifests itself in a large additional low-energy shift of the bound-exciton luminescence peak for $x = 0.10$ composition as the temperature is lowered from 10 to 2K. Furthermore, a significant apparent linewidth broadening was measured. In the interpretation of Ref. 1 this behavior reflects the transition of the bound exciton-spin complex from the thermodynamic-fluctuation-dominated regime to the bound-polaron regime. In the work reported here, we show evidence for the formation of a BMP associated with the bound exciton and connect the measured formation time to the Mn^{2+} -ion spin-spin relaxation processes.

The experimental arrangement employed to perform time-resolved spectroscopy is based on the use of two synchronously pumped, wavelength-tunable dye lasers in an excite-probe configuration, as detailed elsewhere.² In the present instance, a picosecond pulse of excitation promotes a low density of bound excitons (on the order of $1 \times 10^{14} \text{cm}^{-3}$). The presence of this excitation is measured by a weak time-delayed probe pulse as an induced change in its transmission $[\Delta T(\omega)/T(\omega)]$ through the illuminated sample volume.

Under circumstances considered here and for $\Delta T > 0$, the dominant contribution to such optically modulated probe signals is directly proportional to the number of donor sites occupied by the excitons at a give instant of time and energy. This follows by noting that

$$\Delta T/T \sim -\Delta\alpha l \sim \Delta N_{\text{ex}} \times f(\omega)l$$

where ΔN_{ex} is the exciton density, $f(\omega)$ a line-shape function, and l the sample thickness.

Figure 1 summarizes the main features in our data at $T = 2$ K and shows modulated problem transmission spectra recorded at different delays (following the excitation at $t = 0$) at a photon energy of $\hbar\omega_{\text{ex}} = 1.968$ eV, somewhat below the free-exciton energy. The spectra have been properly normalized by accounting for the variation in the absorption coefficient, void of any fine structure.³ By making comparisons with steady-state luminescence spectra,¹ observations from our earlier time-resolved work at higher temperatures, and particularly the appearance of a distinct peak in the *excitation* spectrum at short time delays, we can unambiguously assign the spectral features shown to a bound-exciton complex. The initial formation of a bound exciton for such an excitation energy was confirmed to be rapid (≤ 20 psec) on the time scale of interest here. Most importantly, the data in Fig. 1 show a clear *time-dependent* shift in the probe spectra. In remarkable contrast, we found that the time-resolved spectra observed at 10 K and above (as illustrated in Ref. 3 exhibited *no* measurable shifts but merely a time-dependent amplitude, consistent with the formation and decay of an energetically stationary bound exciton. This constant spectral position of the peak of the modulated transmission at 10 K is shown as the dashed line in Fig. 1. At 2 K we measure an overall red shift in excess of 10 meV within the 800-psec range of our experimental setup. Figure 2 shows the position of

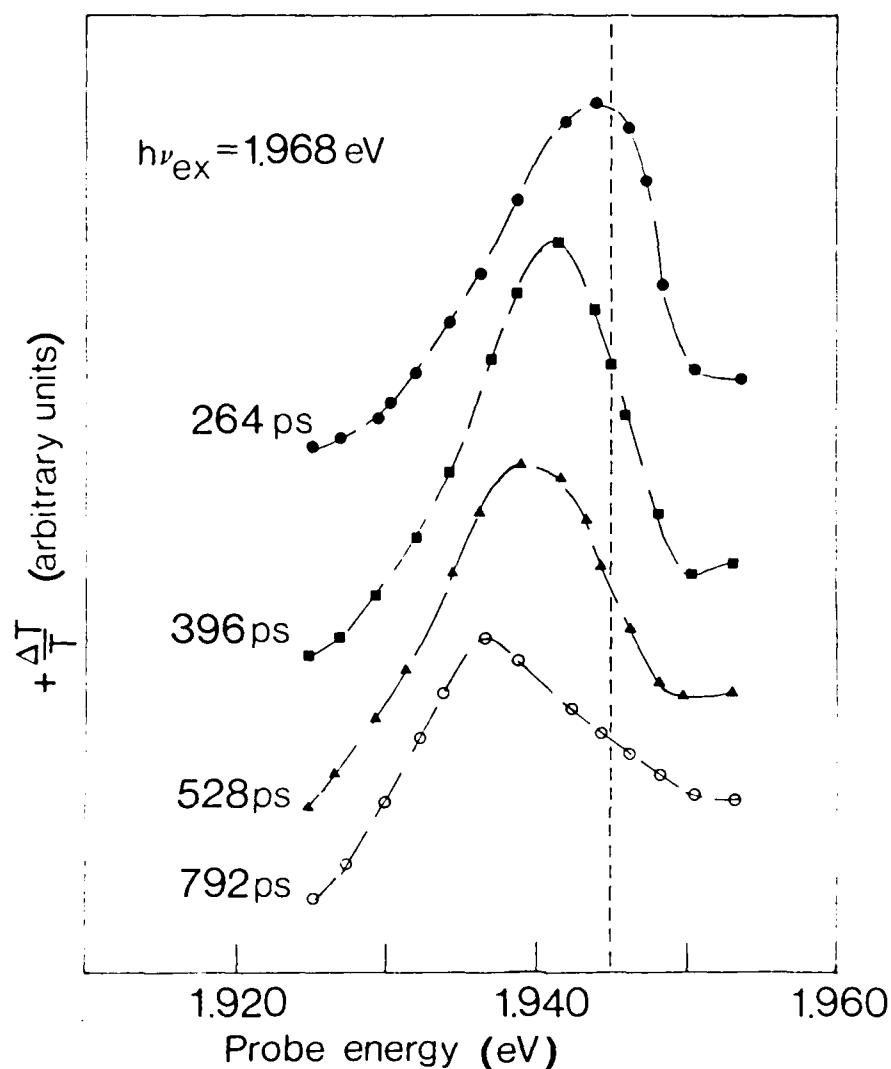


Figure 1: Modulated transmission spectra for the bound exciton in $\text{Cd}_{0.90}\text{Mn}_{0.10}\text{Se}$ at 2K as a function of excite-probe delay in picoseconds (excitation at 1.968 eV). Interpolation of data appears as the dashed line. Relative amplitudes are not fully normalized in this representation. Vertical dashed line denotes the position of the spectrally stationary peak at T= 10K.

the probe spectral peaks as a function of the delay. A good fit to the data can be made with a single exponential time constant of 400 psec. At the same time, the measured single-amplitude decay indicates a lifetime for the bound exciton complex of some 600 psec. The latter is longer by approximately a factor of 3 than a similarly measured lifetime at 10 K.

An additional crucial observation is related to the dependence of the probe spectra on the excitation energy. At all temperatures monitored, a resonancelike feature appeared in the excitation spectrum (as shown at 10 K in Ref. 10). At 2 K, this enhancement had an approximate low-energy limit at about 1.945 eV, below which the probe spectra of Fig. 1 decreased precipitously in amplitude. This provides direct evidence that the time-dependent behavior in Fig. 1 involves a dynamic self-energy relaxation of a bound-exciton state *only initially* accessible for excitation. Some small-amplitude probe signals could be seen when exciting at further lower energies but these contributed to much broader spectra which were also temporally distinctly separate from the bound-exciton kinetics. They appear likely to be connected to localization phenomena from alloy potential fluctuations, coupled with other impurity-related effects probably responsible also for the observed absorption-edge broadening.

We believe that the above observations provide strong evidence for the existence of a bound magnetic polaron in a semiconductor from a direct measurement of its dynamics of formation. The time-dependent Stokes shifts evident in Fig. 1 are here interpreted as corresponding to the different stages of the evolution toward spin ordering about the impurity sites. The occurrence of "polaronic relaxation" is also clearly indicated by the experimental ability to couple excitation directly only to the initial "bare" (spin-uncorrelated) bound-exciton state. When comparing the steady-state photoluminescence spectra

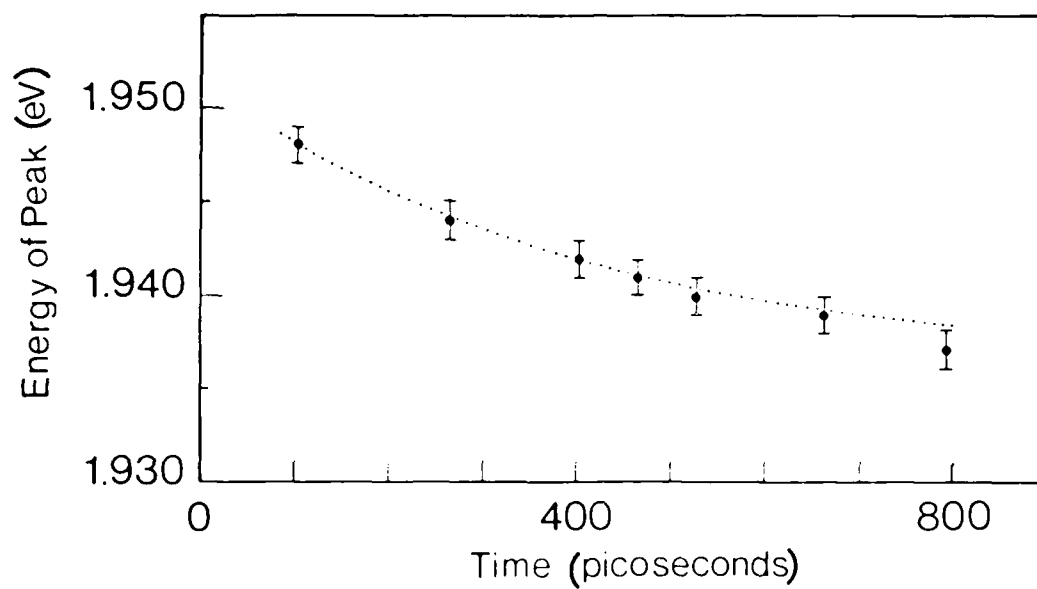


Figure 2: Energetic position of the peak of the exciton line as a function of time delay. Excitation energy in each case is 1.968 eV. Dotted line shows a simple exponential fit.

of Huber *et al.*¹ at 2 K with those in Fig. 1, we find good agreement with their results when time integrating our spectra. This explains the puzzle connected with the anomalously broad linewidths reported in Ref. 1. Furthermore, at a higher temperature of 10 K the transient (instantaneous) and steady-state spectra are in mutual agreement while showing frustration of full polaron formation due to thermal disorder.

Dynamically, the BMP formation can be viewed as an evolution of the bound exciton from an initial statistical regime where a thermally fluctuating individual Mn^{2+} -ion spin has not correlation with the charge carrier spin. To first order the carrier here is represented by the outer hole of the bound exciton. (Our experiment is sensitive to such fluctuations since a large number of laser pulses are used to accumulate each data point.) An inhomogeneously broadened optical linewidth, associated with transitions originating from a distribution of perturbed and spin-split levels is then expected from alloy compositional fluctuations.¹ In the transient experiment such an initial state is prepared by the excitation from an applied laser pulse. At 2 K, the exchange interaction between the carrier spin and those of the ion leads to a mutually correlated spin alignment, the BMP, which in the notation of Ref. 5 can be expressed as an effective magnetization of the form $M(r) = n \exp(-2r/a_x)$, where n is a variationally determined amplitude and a_x the self-consistent Bohr radius of the carrier (in the hydrogenic envelope approximation). The dynamic evolution of the spectra of Fig. 1 is a direct measurement of the development of such magnetization. In the one-electron (hole) model the mean energy shift E_m associated with the lower spin-split level of the exciton can be calculated from the model of Dietel and Spalek in the BMP (mean-field) limit as⁴:

$$E_M = \beta x \tanh(E_M/2kT) / (32\pi g^2 \mu_B a_x^3) \quad , \quad (1)$$

where β is the exchange constant for the outer hole, χ the susceptibility and a_x the Bohr radius. Quantitatively, we find a reasonable agreement of our data with the predicted energy shifts for the formation of the BMP in $\text{Cd}_{0.90}\text{Mn}_{0.10}\text{Se}$. More specifically the BMP contribution adds approximately 15 meV to the exciton binding energy for an assumed Bohr radius of 30 Å by using a exchange constant of $-\beta N_0 = 1.15 \text{ eV}^5$ where N_0 is the number of cations per unit volume. Here we assume that the final state associated with the probe laser transition, i.e., the neutral donor, experiences a much smaller energy shift as shown earlier.¹ In the BMP regime, the alloy compositional fluctuations are still expected to contribute to inhomogeneous linewidths which remain rather broad. One potential complication affecting these estimates concerns the possible participation of the inner pair of electrons on the exchange interactions. Conceivably, the large effective internal magnetic fields (well in excess of 100kG) could alter the bound exciton configuration from that assumed by us.

In conclusion, we have obtained strong evidence from direct time-resolved measurements for the evolution of a photoinduced BMP associated with an impurity bound exciton in $\text{Cd}_{0.90}\text{Mn}_{0.10}\text{Se}$. These results can be connected to previously measured cw photoluminescence spectra which provide a time-integrated contrast to our spectra. While reasonable quantitative agreement exists between experiment and estimates for the BMP energy and formation time in a simple one-particle approach, questions remain concerning the exciton configuration in the BMP limit and the contribution to its formation time by collective spin effects.

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(c) On the Question of Exciton Localization in (Cd,Mn)Te by Alloy Disorder

Many of the recent studies of optical properties in the II-VI diluted magnetic semiconductors (DMS) have involved near bandgap electronic excitations such as free and impurity bound excitons. At the same time there is an expected contribution to band edge broadening from alloy potential fluctuations which are inherent to any mixed crystal due to compositional disorder on microscopic scale. In particular, it has been realized some time ago that excitons may localize in such random potential wells at low temperatures (1). Experimentally, there have been reports in several mixed crystals of evidence for such localization, obtained through analysis of photoluminescence spectra (2), including recent transient spectroscopy (3).

In this segment of the accomplishments under the AFOSR supported research, we highlight results of time-resolved studies on subnanosecond timescale of excitonic spectra in $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ ($x=.15$) where picosecond laser techniques have been applied to look for evidence of exciton localization. The results are discussed below only in a qualitative way. Golnik and Lavallard have also performed similar work recently (4). Since $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ grows usually slightly p-type but with substantial compensation, one expects (neutral) acceptors and alloy fluctuations to compete with each other in the capture of free excitons injected to a sample. At higher impurity concentrations (but below the Mott transition) the formation of impurity bound excitons becomes the most likely event, as shown by us recently in n- $\text{Cd}_{1-x}\text{Mn}_x\text{Se}$ through time resolved spectroscopy

with the donor bound exciton (5). Localization by alloy potential fluctuations might take on additional significance in the DMS because of the possibility of a subsequent polaronic effect from the exchange interaction between the spins of the carriers and localized Mn-ion magnetic moments. Much of the recent experimental evidence for the 'bound magnetic polaron' (BMP) has come from studies of impurity bound exciton spectra (6). In that case, the electrostatic binding of a free carrier or a free exciton to an impurity complex provides the necessary initial confinement of the quasiparticle(s) in an effective Bohr volume.

Our experiments employed two picosecond dye lasers to excite and probe the $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ samples. We have used such photomodulation spectroscopy in several earlier occasions involving free and impurity bound excitons in semiconductors (7). One advantage of this approach over time resolved luminescence is the ability to distinguish spectral signatures between free and bound (localized) excitons. This follows from the distinct lineshape contribution which is made by exciton-exciton scattering to collisional broadening in such spectra for free excitons. Another benefit is the ability to apply resonant excitation without interference by scattered pump light in a noncollinear geometry with the help of suitable electro-optical modulation techniques (8).

In this work we concentrated selectively on $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ samples ($x=.15$) where cw-photoluminescence spectra did not show the strong emission characteristic of the neutral acceptor bound exciton (A^0X). (In samples where such A^0X emission dominates, we saw the large temperature dependent shifts in the emission energy, associated earlier with the BMP effect by Golnik et. al. (5).) Figure 1 shows luminescence spectra at three

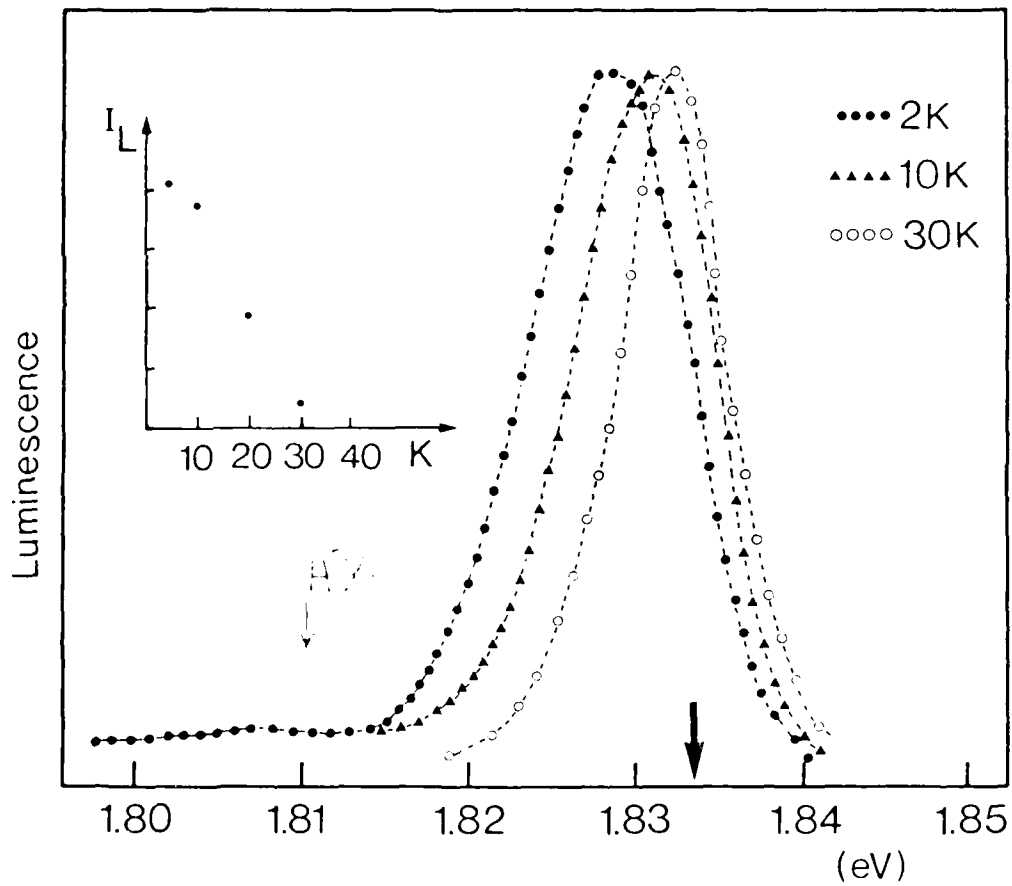


Figure 1: Photoluminescence in $(\text{Cd,Mn})\text{Te}$ ($x = 0.15$) at three different temperatures in the absence of a strong $\text{A}^{\text{O}}\text{X}$ emission. The arrow indicates the position of the free exciton as determined from reflectance at $T = 2\text{K}$. The inset shows the actual temperature dependence of the luminescence amplitudes.

different temperatures, where the peaks are typically 10 meV and more above the known A^0X energies. The free exciton energy as determined from reflectance at $T=2K$ is also indicated, together with the temperature dependence of the amplitude. Note the blueshift of the peak energies with temperature and the narrowing of the spectra (the shift is, however, considerably less than that observed with the A^0X emission). By $T=30K$ the high energy side of the spectrum has a width comparable to kT . The spectra could suggest a (direct gap) material where close compensation has sufficiently reduced the density of neutral acceptor sites so that low temperature free exciton localization to near band edge states may have become possible. At the same time, nonexcitonic emission from other shallow impurity states is likely to add a contribution.

To test this possibility further, Figure 2 shows transient photomodulated spectra in this energy region, initiated by picosecond laser excitation in the same samples at $T=2K$. The absorbed photon density per pulse was maintained below 10^{15} cm^{-3} . The photon energy of excitation in Fig. 2a corresponds approximately that of the free exciton (from reflectance spectra) and that in Fig. 2b lies below it. Analysis of the photomodulation approach shows that the spectra are qualitatively those expected for a noninteracting exciton gas (in nonextended states), and can then be thought as analogous to time resolved photoluminescence. We see a pronounced difference between Figures 2a and 2b, with the rather narrow and distinct peak appearing at the photon energy of excitation in the latter. For photon energies of excitation varying over a range of nearly 10 meV below the free exciton energy such a peak was evident, tracking well the energy of excitation. At delay times $t > 0$ a distinct low energy shoulder appeared, evolving while its spectral center of gravity moved to

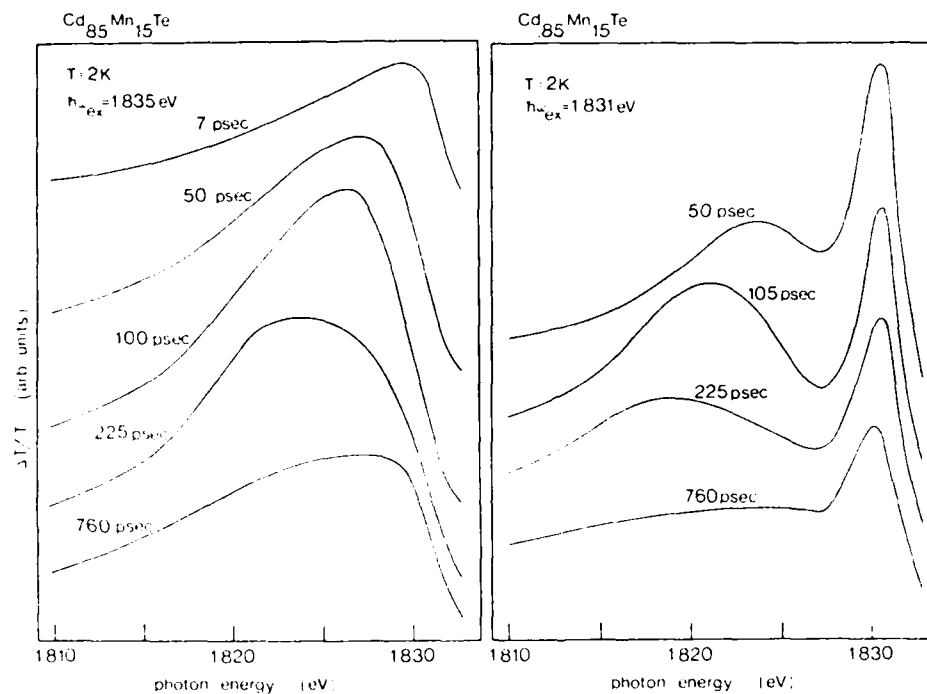


Figure 2: Transient spectra in $x = 0.15$ material at $T = 2$ K below free exciton energy for two different energies of excitation: (i) at the free exciton energy (2a; left), and below the free exciton (2b; right).

lower energies. A time dependent shift is also seen in the broader nonresonantly excited features of Fig. 2a, following the free exciton injection at $t=0$. In all cases we also saw a long lived spectrally stationary 'background' contribution which is apparent at the longest graphed delay times in Fig. 2. The lifetime associated with this background was typically many nanoseconds.

With increasing temperature, the peaks and their associated shoulders such as in Fig. 2b lost their distinctive shapes as well as decreased in amplitude when sample temperatures reached $T=10$ K. This change was also clearly seen through an increasingly rapid decay of the 'resonance' peaks (e.g. about 100 psec at $T=5$ K for the peak in Fig. 2b). At the same time, the long lived 'background' signals became relatively more dominant in this energy region, while at higher photon energies we saw spectral features which were suggestive of free exciton like behavior. The free exciton photomodulation spectra is usually distinguishable from the appearance of a sign change in the experimentally measured quantity dT/T originating from collision broadening of the exciton line (9). In our samples we also measured a substantial redshift in the tail of the absorption edge with increasing temperature. (For reference, at a value 100 cm^{-1} for the absorption coefficient, the redshift of the edge had an average value of 1.5 meV/K in the range of 2 to 20 K).

We interpret these results in the following way, while using qualitatively many of the ideas of Cohen and Sturge in Ref. 2. At the low temperatures, data such as in Fig. 2 is taken to imply the direct observation of localization of free excitons in alloy potential fluctuations. We can crudely estimate (10) the range of energies for such

localized states below the free exciton edge to be approximately 10 meV, if the Mn-ion distribution is statistical (there is uncertainty at present about the degree of clustering which may take place in this alloy). The relatively sharp structure which can be resonantly excited (Fig. 2b), is central to this argument, together with its disappearance at higher temperatures. The absence of a lower energy shoulder at $t=0$ and its appearance only at $t>0$ is interpreted as originating from acoustic phonon assisted tunneling to lower energy states in the continuum distribution of localized states (the details of such density of states are difficult to address from our data). An energy dependent tunneling cross-section together with a particular density of states can give rise to the appearance of a distinct shoulder as calculated in more detail by Cohen and Sturge (2).

In this interpretation, nonresonant excitation to the free exciton states above a 'mobility edge' (Fig. 2a) is followed by an initial capture to most of the localized states and subsequent energy relaxation within these states. From the time dependent data we infer capture rates typically on the order of 10^{11} sec^{-1} at $T=2 \text{ K}$, slower than those seen earlier with impurity bound exciton formation (5). The low lattice temperature is unlikely to permit a significant thermal ionization and thus a multiple trapping like relaxation process should be less probable than the phonon assisted tunneling. From the observed spectral diffusion we extract tunneling rates which are on the order of 10^9 sec^{-1} at $T=2 \text{ K}$, i.e. comparable to the exciton lifetime (among the approximations made in this estimate is that of a constant optical matrix element within the localized states). Thus, a complete thermalization would not be achieved. At higher temperatures, the role of multiple trapping is expected to

increase the thermal equilibration rate within the localized states as is qualitatively seen in our data. The amplitude of the cw-photoluminescence signal decreases rapidly at temperatures above 10 K suggesting a nonradiative trapping mechanism for free excitons. This behavior has also been confirmed earlier by us in time resolved experiments for the free exciton in $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ (11).

The evidence that free exciton localization is being directly seen in these experiments cannot be completely separated from the unavoidable effects of impurities and other defects in the presently available $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$. In particular, assuming a nearly compensated p-type (high resistivity) material implies that the position of the equilibrium Fermi level lies in the vicinity of the valence band edge at low temperatures. In fact, we suggest that E_F overlaps the valence bandtails which originate from the compositional disorder (the usual impurity/vacancy acceptor energy in CdTe is somewhat larger than the expected band edge smearing). With increasing temperature some thermal ionization of electrons into the unoccupied localized states takes place thus shifting E_F deeper into the gap and giving rise to the observed shift in the tail of the absorption edge. This also makes possible single electron (nonexcitonic) contributions in the photomodulated spectra, which follow the excitation of an electron from a defect state into the conduction band tail states. Any possible subsequent single carrier localization would be expected to result in longer lifetimes and is likely to be associated with the 'background' signal spectra observed by us.

In summary, we have presented experimental results and arguments that free exciton localization in alloy potential fluctuations can take place

in $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ when the competition by impurity bound exciton formation is not dominant. However, we have not yet seen any magnetic polaron-like effects under conditions of direct excitation to localized states although additional tests using an external magnetic field are under way. We also note that, in general, the real time spectral diffusion observed by us can also be expected from an inhomogeneously broadened impurity band. Finally, we would like to contrast these results with our recent work in $n\text{-Cd}_{1-x}\text{Mn}_x\text{Se}$, where transient spectra associated with neutral donor bound exciton was used to support arguments for the formation of an BMP (7).

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(d) Optical Properties of a New Infrared Material: $\text{Pb}_{1-x}\text{Eu}_x\text{Te}$

In this phase of the AFOSR supported research we have focussed on optical studies of a novel semiconductor material, $\text{Pb}_{1-x}\text{Eu}_x\text{Te}$. Recent advances in molecular beam epitaxial growth of $\text{Pb}_{1-x}\text{Eu}_x\text{Te}$ have demonstrated the usefulness of this narrow gap material for infrared optoelectronic applications, specifically in the fabrication of $\text{PbTe}/\text{Pb}_{1-x}\text{Eu}_x\text{Te}$ heterojunction diode lasers including quantum well structures (1),(2). The range of Eu-concentrations so far employed in these devices has been relatively low ($x < 0.05$) and useful information has been obtained about the basic optical properties of $\text{Pb}_{1-x}\text{Eu}_x\text{Te}$ and the $\text{PbTe}/\text{Pb}_{1-x}\text{Eu}_x\text{Te}$ multiple quantum wells (MQW) in this concentration range (3),(4). On the other hand, very little is presently known about the basic properties of $\text{Pb}_{1-x}\text{Eu}_x\text{Te}$ for higher Eu-concentrations, limited to an early study on evaporated thin films (5). Among the important issues for experimental measurement is the concentration dependence of the effective (optical) energy gap and associated magneto-optical effects. While for low x -values the bandedge states of $\text{Pb}_{1-x}\text{Eu}_x\text{Te}$ are expected to be PbTe-like (composed primarily of p-orbitals with a direct gap at the L-point in the Brillouin zone), experimentally derived arguments for the bandstructure of the antiferromagnetic EuTe show the importance of the Eu^{2+} -ion f- and d-electron states in defining the optical gap (6). Since Eu enters the alloy as a divalent ion, the half-filled f-electron shell is expected to contribute a magnetic moment from a pure spin state ($S=7/2$ in the free ion). This raises the question of the spin exchange interaction between the Bloch states and the ground f-electron states as already investigated broadly in the II-VI 'semimagnetic' semiconductors as well as in $(\text{Pb},\text{Mn})\text{Te}$ and $(\text{Pb},\text{Mn})\text{S}$. In this section of our report we present results of

luminescence and absorption studies on several thin film samples over a concentration range up to $x=0.30$, which have allowed us to speculate about the nature of the electronic states which define the effective optical bandgap in this alloy. In addition, we also discuss measurements of the magnetic field dependence of the optical gap on selected samples which we use for an empirical description of the 'p-f' spin exchange effects in $\text{Pb}_{1-x}\text{Eu}_x\text{Te}$ at moderate Eu-concentrations ($x < 0.20$). We also briefly note observations on samples of higher Eu-concentration.

The samples used in these studies were MBE-grown single crystal thin films of $\text{Pb}_{1-x}\text{Eu}_x\text{Te}$ on (111) oriented BaF_2 substrates (by Dr. D.L. Partin at General Motors Research Laboratories). No effort was made to control the strain induced by the finite film/substrate lattice mismatch although lattice matching to PbTe substrates with the addition of selenium at low x -values has been demonstrated (1). The thin films varied in thickness from approximately 0.3 μm to 30 μm and were, in most cases, not intentionally doped. However, nonstoichiometric growth under conditions of excess Te-vapor leads to p-type material with a hole density on the order of $1-2 \times 10^{17} \text{ cm}^{-3}$.

Low temperature photoluminescence measurements were performed with a continuous wave Nd:YAG laser as the excitation source ($\hbar\omega = 1.17 \text{ eV}$) at low excitation intensities. The samples were mounted on the coldfinger of a standard variable temperature dewar for both luminescence and absorption measurements, while a separate superconducting magnet dewar was used for the field dependent studies. Figure 1 shows luminescence spectra for two $\text{Pb}_{1-x}\text{Eu}_x\text{Te}$ films of $x=0.032$ and 0.20 at $T=10\text{K}$ (see comments below about the concentrations). The linewidth of the sample of lower Eu-concentration

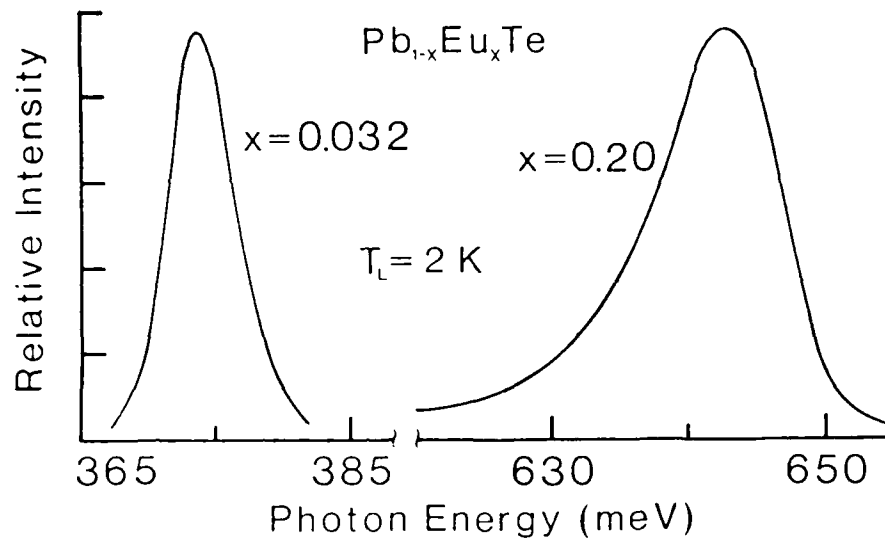


Figure 1: Photoluminescence spectra at the optical gap from two MBE grown thin film samples of $(\text{Pb},\text{Eu})\text{Te}$ at $T = 2\text{ K}$.

(left panel) is typical for a low density electron-hole plasma recombination spectrum. The sample with $x=0.20$ shows a pronounced broadening on the low energy side, similar to that observed in other semiconducting alloys at increased concentrations and which has been modelled in terms of alloy potential fluctuations (7).

The optical bandgap as a function of Eu-concentration obtained from both luminescence and absorption measurements on a number of $\text{Pb}_{1-x}\text{Eu}_x\text{Te}$ thin films up to $x=0.30$ is shown in Figure 2 at $T=10\text{K}$. The luminescence spectra frequently showed additional emission features at lower photon energies and we studied such spectra carefully (e.g. as a function of excitation density) before extracting values for the optical gaps. The effective gap energies were then defined from the peak photon energies for the luminescence spectra. The absorption data was used to correlate this information; in particular for $x>0.05$ corresponding values for the optical gap were obtained from a linear extrapolation of $(\text{absorbance})^2$ against the photon energy axis. The choice of this parabolic functional form has no rigorous justification in this case because of the unknown energy dependences of the densities of states, and it was simply chosen as a best fit over a wide concentration range. Using the relatively narrow luminescence linewidths and the rather good linear fits to $(\text{absorbance})^2$ vs. photon energy data (over a limited energy range), the two procedures generally agreed to within 5-10 meV on the several samples for which such consistency was checked. The dashed line in the figure refers to the recently derived dependence of $E_g(x)$ at low concentrations ($x<0.04$), obtained from emission energies of $\text{Pb}_{1-x}\text{Eu}_x\text{Te}$ lasers (1). Furthermore, we verified that up to 77K the empirical temperature dependent expression for $E_g(x,T)$ of Ref. 2 was obeyed rather well. However, we also accounted for

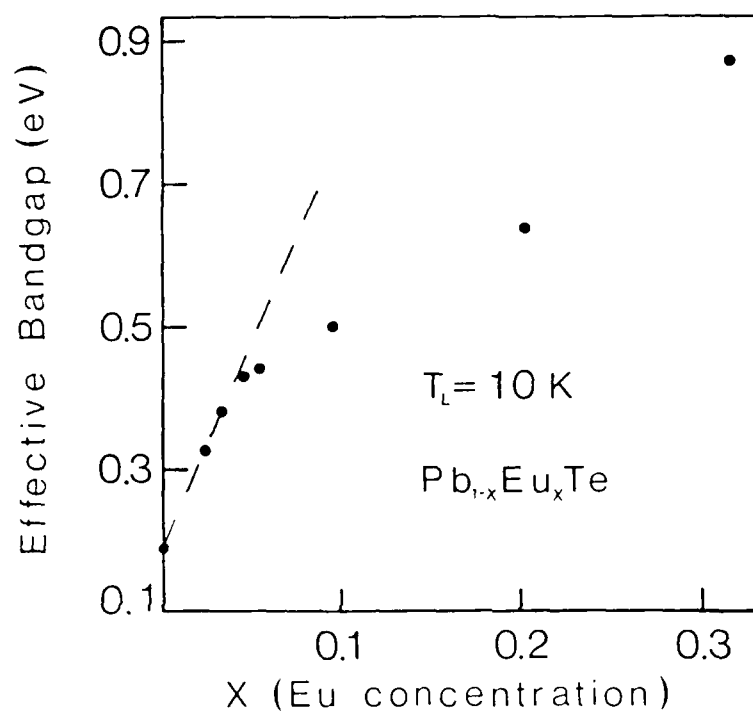


Figure 2: Optical gap of $(\text{Pb},\text{Eu})\text{Te}$ as a function of the Eu-concentration at $T = 10\text{ K}$ as obtained from luminescence and absorption measurements. The dashed line refers to the empirical expression in Ref. 3.

the finite uncertainties in the composition and strain in our samples by explicitly assuming that the dependence of E_g on x is indeed linear in this range. Our data in Figure 2 then clearly shows that while the optical gap opens up rapidly for low x -values ($dE_g/dx = 5.788$ eV for the dashed line), the slope decreases strongly for $x > 0.05$. Furthermore, while for samples with x up to 0.10 the thermal coefficient $dE_g/dT > 0$, we found that this coefficient had changed sign for the $x=0.20$ sample (and higher values). Results for three other thin film samples at considerably higher concentrations $x > 0.3$ ($x=0.47, 0.71, 1.00$) showed evidence that the electronic character of the states defining the optical gap (in absorption) is now further strongly changing (with x) from a PbTe-like system. Among other things, this manifests itself with an increasing Stokes shift between the luminescence and absorption edges, reaching a value of approximately 0.7 eV in the EuTe limit, as will be detailed elsewhere (8).

Magnetic field dependence of the bandedge luminescence spectra for the two $\text{Pb}_{1-x}\text{Eu}_x\text{Te}$ thin film samples discussed above in connection with Fig. 1 is summarized in Figure 3, where the peak emission energies are plotted up to a field of 5 Tesla in the Faraday geometry. Note that while the sample with $x=0.032$ shows predominantly a spectral blueshift with increasing field, the peaks for the $x=0.20$ thin film display a strongly temperature dependent spectral redshift. The dashed lines correspond to a model calculation discussed below.

We now consider these measurements and their implications on the electronic character of those states which define the effective bandgap in $\text{Pb}_{1-x}\text{Eu}_x\text{Te}$. Our observations are largely empirical in nature and have

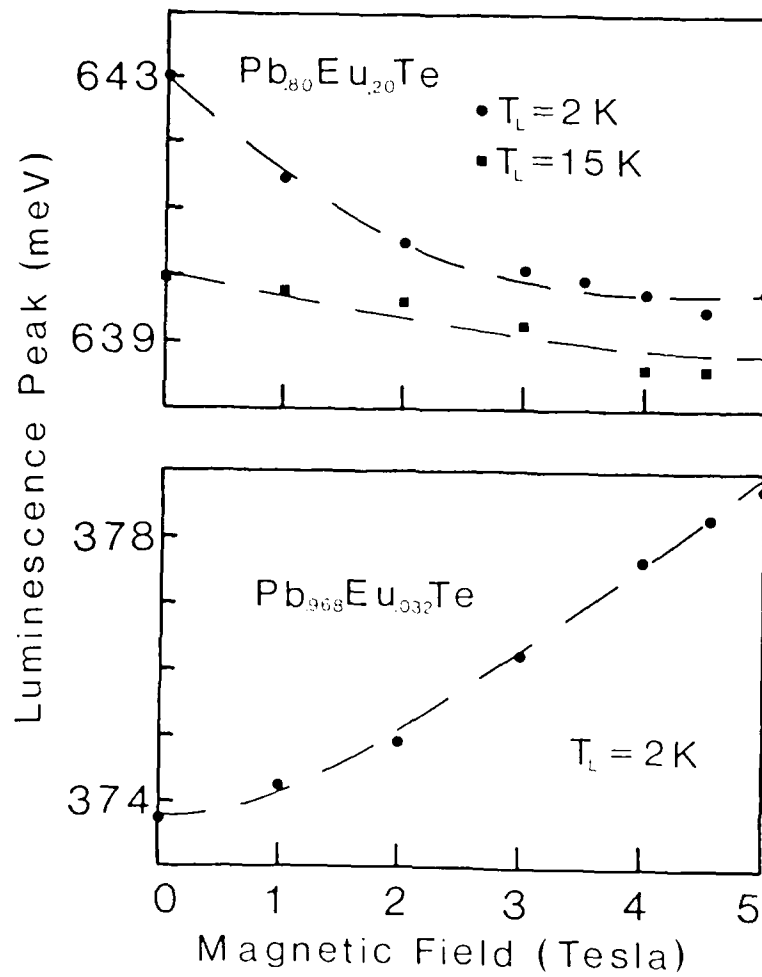


Figure 3: Peak position of the luminescence emission (in meV) as a function of external magnetic field for two samples of different Eu-concentration. The dashed lines refer to the model calculation discussed in text.

partial difficulty in the fact that the bandstructure for EuTe appears still to be under some theoretical debate. At low x -values ($x < 0.05$), however, it seems clear that the effective gap is composed of conduction and valence band states which are still predominantly 'PbTe-like'. The large slope dE_g/dx is assumed to be constant and $dE_g/dT > 0$. The magnetic field induced shifts in the luminescence emission energy (lower panel in Fig.3) are compatible with optical transitions which occur between the lowest spin split Landau levels of the conduction and valence bands. A smaller (oppositely shifting) contribution from an exchange term is also present as summarized below. The striking feature in Fig. 1 is, of course, the rather precipitous decrease in the slope dE_g/dx for $x > 0.05$. With increasing Eu-concentration, magneto-optical data for the $x=0.20$ sample and the observation of $dE_g/dT < 0$ support the argument that substantial additional mixing to the initially p-like conduction and valence orbitals is continuing to take place. This hybridization can in principle involve the s-, p-, and f- electron orbitals of the Eu^{2+} -ion but we do not estimate their contributions in this paper any further (in part because of the lack of e.g. useful tight-binding parameters). Finally, the Stokes shifts between the absorption edge and the luminescence spectra which become clearly evident beyond $x > 0.30$ show that in this regime of concentrations, further changes have been introduced by the Eu^{2+} -ion to those states which define the optical gap (in absorption). Specifically, the Stokes shifts appear to reflect a strong configurational relaxation (Jahn-Teller) and thus point towards the introduction of the f-states into the gap.

To characterize the magneto-optical effects shown in Figure 3 further, we have taken a first order phenomenological approach to test one possible

description for the exchange interaction between localized paramagnetic Eu^{2+} -ion moments with spins of extended electronic states (which lead to spin splittings of the conduction and valence band states). This approach is well established in the wider gap II-VI compound 'semimagnetic' semiconductors (with Mn^{2+} as the paramagnetic ion) where the spin exchange is a Heisenberg-like short range interaction (9). Also, in the present case, the relatively low background free carrier concentration makes an RKKY-like exchange mechanism of negligible importance. Accordingly, we have fitted the data in Fig. 3 to the following parametrized expression:

$$\Delta E = \Delta E_0(B, m^*, g) - A(x) \times B(g' \mu_B / k(T + T_0)) \quad (1)$$

Here ΔE is the spectral shift in the luminescence emission energy (optical gap) and the first term on the right, ΔE_0 , is the contribution from the spin-split lowest Landau-level (proportional to the applied external field in the range of interest here). The second term, where the Eu-concentration is x , originates from the assumed Heisenberg-like exchange mechanism with a Brillouin function B in whose argument g' is the g -factor for the Eu^{2+} -ion. As for the free ion, we take $S=7/2$, $L=0$, and assume that Hund's rule is valid. By using the empirical treatment applied earlier to the II-VI semimagnetic semiconductors (10), we account for the finite antiferromagnetic (AF) spin-spin interactions within the Eu^{2+} system by using a temperature correction T_0 in the argument of the Brillouin function so that $T_{\text{eff}} = T + T_0$, and define a concentration dependent interband exchange coefficient $A(x)$. Note that our present experiment is not able to separate out the valence and conduction band contributions to these exchange coefficients. For evaluation of the exchange effect we can subtract away the 'Landau level contribution' to

lowest approximation by extrapolating from measured magneto-optical constants of PbTe (11) in terms of an effective mass (and g-factor) which have been assumed to have an approximately inverse dependence on the effective mass for the different x-values. Then the fits in Figure 3 are obtained for the following parametrized values of the exchange term in Eq. 1: (i) for the sample with $x=0.032$, $A=49$ meV and $T_0 < 0.5$ K; (ii) for $x=0.20$ material $A=30$ meV and $T_0=4.3$ K in the lower temperature case. For the lattice temperature of $T=15$ K we find that the best fit requires a somewhat different (lower) value of $T_0=2$ K; this may imply that the description of Eq. 1 is not quite correct or show imprecision in our treatment of the Landau contribution. We also note that the forefactor $A(x)$ decreases with x, as expected from increasing AF contributions, and that no spin saturation is evident at the moderate magnetic fields and temperatures employed in these experiments.

These estimates, while based on a limited amount of information and simplified in character, do suggest that even at $x=0.20$, the conduction and valence band extrema of (Pb,Eu)Te are still largely composed of extended states. The approximate fitting to a Heisenberg-like spin exchange formulation to account for the magneto-optical shifts is not necessarily a unique solution to the problem but seems to us to be the most likely one. We have some support to these estimates from recent magnetization (and susceptibility) measurements which have yielded more direct information e.g. about the role of the AF interactions (12). These measurements on similar MBE grown (Pb,Eu)Te thin film samples show, for example, that the Eu^{2+} -spin system is paramagnetic over the range of compositions ($x < 0.20$) and temperatures considered here (12). Furthermore, fitting the magnetization to the modified Brillouin function yields

temperature corrections T_0 which are in good agreement with the values obtained above. We note in passing other recent susceptibility measurements for (Pb,Eu)Te in a low concentration range (x from 0.001 to 0.018) which show very large (negative) Curie-Weiss temperatures (13). This has been interpreted in terms of the long range exchange interaction in a narrow-gap system such as (Pb,Eu)Te; however, the concentration range discussed in this paper is much higher and is likely to involve more direct processes. Finally, using the values extracted for the exchange amplitudes $A(x)$ from above, we note that these are between one and two orders of magnitude smaller than the corresponding joint conduction-valence band contributions of exchange in the wider gap II-VI semimagnetic semiconductors. At the same time, magneto-optical effects in the narrow gap materials $\text{Pb}_{1-x}\text{Mn}_x$ and $\text{Pb}_{1-x}\text{Mn}_x\text{S}$ have also been measured to be quite small (14), (15).

In summary, we have obtained through infrared luminescence and absorption measurements information about the optical bandgap of $\text{Pb}_{1-x}\text{Eu}_x\text{Te}$ over a wide range of composition. While using additional data from magneto-optical luminescence studies we suggest that in this narrow-gap mixed crystal, the character of the states which define the effective energy gap remains largely extended at least up to $x=0.20$. The origin of the pronounced change in the slope of dE_g/dx for $x>0.05$ is clearly a major unresolved theoretical problem in this connection, and apart from questions of hybridization, may also have a magnetic contribution through coupling with the Eu^{2+} -ion magnetic moments. The magneto-optical effects at the optical gap have been found to be rather small but seem at least approximately fit to a simple exchange description. Here also a proper theory will require systematic additional

experimental input as a function of concentration x , including detailed correlation between magneto-optical and susceptibility data. Finally, approximately beyond $x=0.30$ the effective bandgap in $(\text{Pb},\text{Eu})\text{Te}$ appears to contain states which are now directly related to the Eu^{2+} -ion f-electron (and possibly d-electron) bands.

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(e) Electron-Hole Recombination Spectra and Kinetics in PbTe/PbEuTeSe

Multiple Quantum Wells

In this segment of the AFOSR supported research we have focussed on the optical properties of electronic excitations in new narrow-gap semiconductor ultrathin layer structures, i.e. quantum wells, based on the $\text{Pb}_{1-x}\text{Eu}_x$ material system. As an example we present here studies of PbTe/PbEuTeSe multiple quantum wells in samples of two different well thicknesses ($L_z=92$ Å and 51 Å) from measurements of photoluminescence over a range of temperatures and magnetic fields structures grown by Dr. D.L. Partin at General Motors Research Laboratories). From infrared luminescence we obtain transition energies which yield tentative assignments for values of the band offsets. Hot electron effects are pronounced in the $L_z=92$ Å sample and show that the electron-LO phonon interaction is substantially reduced in comparison with bulk PbTe. The energies of the lowest Landau levels have been measured in several samples from shifts in luminescence energies in external fields. Finally, measurements of excess electron-hole recombination rates by time-resolved techniques show lifetimes less than 10 nsec in these quantum wells.

The PbTe/PbEuTeSe multiple quantum well structure has been successfully fabricated for diode laser operation recently (1) and presents a useful system for study of quasi-2D electronic excitations through interband optical transitions because of the very small excitonic corrections. We have investigated basic optical properties of this superlattice for narrow wells ($L_z < 100$ Å) through photoluminescence measurements as a function of incident excitation level, temperature and external magnetic field. With pulsed laser techniques we have also measured the recombination lifetimes and find them typically to be less than 10 nsec at temperatures below 77 K.

The samples used in this study were grown by MBE techniques on (100) oriented PbTe substrates (2). Buffer layers of PbEuSeTe of thicknesses 1-2 microns separated the quantum well layers from the substrate. Below, we show results for two samples, a PbTe/Pb_{1-x}Eu_xTe_{1-y}Se_y structure with PbTe well thickness $L_z = 92$ Å and barrier thickness $L_b = 940$ Å, and a structure with $L_z = 51$ Å and $L_b = 355$ Å. The Eu concentrations for the two samples were $x = .048$ and $x = .040$, respectively. The selenium concentrations ($y = .059$ and $.046$) were chosen to minimize lattice mismatch and are not expected to appreciably change the near bandgap electronic properties of the ternary PbEuTe. The PbTe layers were assumed to have a background hole concentration of about $2 \times 10^{17} \text{ cm}^{-3}$ from comparison with data obtained for single crystal films of PbTe grown under similar conditions.

Figure 1 shows infrared luminescence spectra for two MQW samples at a nominal lattice temperature $T = 4\text{K}$, obtained by photoexcitation from a Nd:YAG laser ($\hbar\omega = 1.17$ eV) under moderate power ($P < 50$ mW). For the wider well sample ($L_z = 92$ Å) the spectrum consists of a single feature ($n=1$ to $n=1$ conduction to

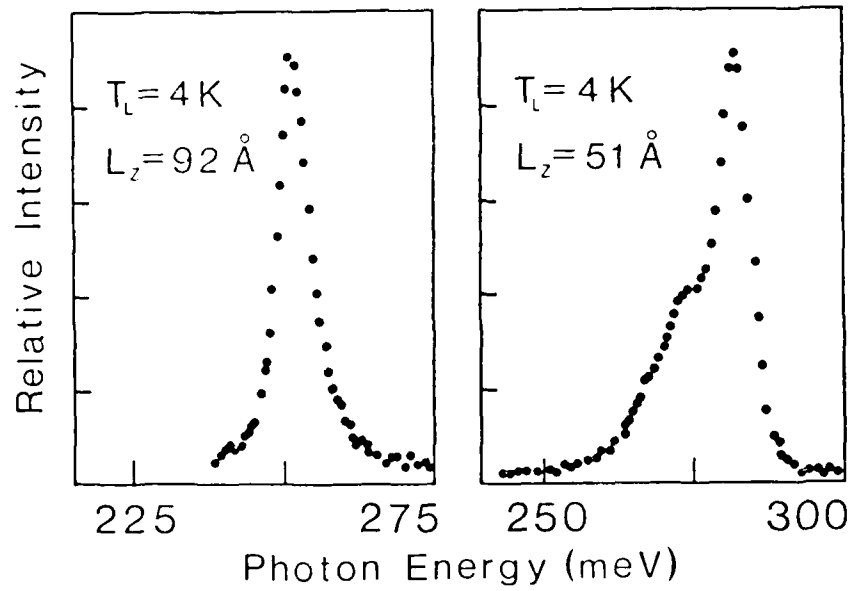


Figure 1: Photoluminescence from two PbTe/PbEuTeSe multiple quantum wells at $T=4\text{ K}$ under moderate cw excitation. The amplitudes for each spectra are normalized to their respective peaks.

valence band transition) which acquires a nearly Boltzmann like tail at higher temperatures and excitation levels. We have shown recently how the excitation intensity dependence of the spectra for such MQW samples leads to readily observable hot carrier effects which have been interpreted in terms of a significantly reduced electron-optical phonon interaction of the photoexcited carriers (3). Furthermore for these samples, the low energy portion of the lineshape fits well with a model in which well width fluctuations of one monolayer contribute to the observed finite broadening. In contrast, as the well width decreases to approximately 50 Å, a distinct low energy shoulder appears in low temperature luminescence (Fig.1). The shoulder (whose origin is not understood at present) decreases in amplitude and disappears with increasing temperature ($T > 20$ K), but even in its absence only a poor fit can be made to the lineshape by only considering well width fluctuations. Comparison with another MQW sample with comparable dimensions ($L_z = 52$ Å) but grown at a higher temperature (390 °C vs. 320 °C) shows nearly identical spectral behavior; this shows that interdiffusion is not a dominant factor in the thin well samples. From the spectral positions of the luminescence for the two samples in Fig. 1, we estimate that the conduction band offset ratio in the PbTe/PbEuTe heterojunction $\Delta E_c / \Delta E_g = .90$ or $.10$.

The influence of an external magnetic field on the luminescence spectra is shown in Fig. 2 for the 51 Å MQW sample. The lower part of the figure shows the dependence of the spectral position of the main peak at 2K for two orientations of the field vs. the superlattice growth axis z (parallel to [100] direction). The optical transition is assumed to correspond to lowest spin allowed transition between the conduction and valence band quantum well Landau levels. Clear anisotropy in the field induced shifts $\Delta E(B)$ is evident, as expected for a quasi-2D transition involving free electrons and holes. The upper part of the

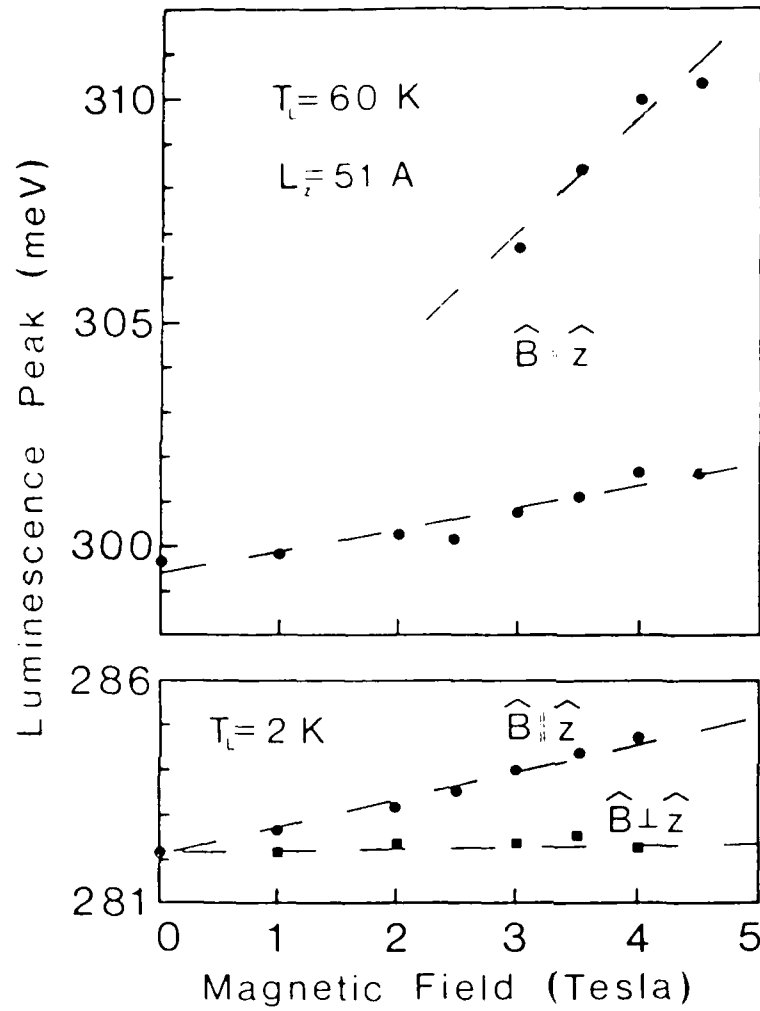


Figure 2: Influence of an external magnetic field on the position of the luminescence peaks for a sample of 51 Å PbTe quantum well width. Lower trace: Dependence on field orientation vs. superlattice axis at $T=2$ K. Upper trace: Lowest resonance and the appearance of the next spin/Landau transition at $T=60$

figure shows the field induced shift at 60K in Faraday geometry where, in addition to the lowest resonance, a next spin/Landau level transition has sufficient thermally excited population to be clearly resolved in the luminescence spectra at higher field values. The dashed straight lines are to guide the eye only. The absence of any strong temperature dependence in the slope $\Delta E/\Delta B$ shows that little contribution is made by any spin exchange effects of the electrons and holes with the Eu-ion f-electrons in the PbEuTe barrier (in spite of the substantial penetration of their wavefunctions into this region). At the same time, comparison with reported interband magneto-optical data in bulk PbTe (4) shows that the field induced shifts in our thin well MQW samples are substantially smaller. Details of a model which accounts for the magneto-optical behavior in the (100) PbTe/PbEuTeSe superlattices will be discussed elsewhere.

Finally, we have also made measurements of the recombination lifetime in several MQW samples by time-resolved photoluminescence. The source of excitation in these experiments was a cw modelocked Nd:YAG laser ($t_p = 100$ psec). At low excitation levels where transient luminescent signals are weak for direct detection, a useful experimental technique is based on a phase-shift approach to characterize the luminescence decays (5). Briefly, the high repetition rate pulsed output (100 MHz) of the laser was additionally amplitude modulated at 10 MHz and the relative phase of the photodetected luminescence from the MQW samples was compared with that directly emitted by the laser through a high frequency lock-in amplifier. As an example, at 77K our measurements indicate approximately an electron-hole lifetime for the $L_z=92$ MQW sample of 6 nsec and for an $L_z=51$ A sample a lifetime of 4 nsec. These lifetimes become somewhat longer towards lower temperatures (to about 4K). Considering the high radiative efficiency and the moderate excitation levels (excess electron density $< 1 \times 10^{17} \text{ cm}^{-3}$) our measurements indicate that a substantial shortening in the radiative

lifetime in the quantum well structures has occurred when compared with reported values in bulk PbTe under comparable conditions (6). Additional experiments are now under way to examine the lifetimes in the PbTe/PbEuTeSe quantum well structures at higher carrier densities in order to examine the characteristics of Auger recombination in the quasi-2D case.

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3. Scientific Publications and Presentations Resulting from the AFOSR supported Research

1. "High Resolution Picosecond Modulation Spectroscopy of Near Interband Resonances in Semiconductors", S. Sugai, J.H. Harris, and A.V. Nurmikko, in Picosecond Phenomena III, Springer Series in Chem. Physics, vol.23, p.103 (1982)
2. "Transient Polarized Modulated Reflection Spectra of Near Bandedge Excitations in $\text{Cd}_{.70}\text{Mn}_{.30}\text{Te}$ ", A.V. Nurmikko, S. Sugai, and J.H. Harris, Proc. Int. Conf. Physics of Semic., Montpellier, Physica B117, 494 (1983)
3. "Magnetic Polaron Contribution to Donor Bound Exciton in $(\text{Cd,Mn})\text{Se}$ ", C.A. Huber, A.V. Nurmikko, M. Gal, and A. Wold, Solid State Comm. 46, 41 (1983)
4. "Picosecond Kinetics of the Donor Bound Exciton in $(\text{Cd,Mn})\text{Se}$ ", J.H. Harris and A.V. Nurmikko, Phys.Rev. B28, 1181 (1983)
5. "Formation of the Bound Magnetic Polaron in $(\text{Cd,Mn})\text{Se}$ ", J.H. Harris and A.V. Nurmikko, Phys.Rev.Lett. 51, 1472 (1983)
6. "Subnanosecond Luminescence Spectroscopy of $(\text{Cd,Mn})\text{Se}$ under High Intensity Optical Excitation", C.A. Huber and A.V. Nurmikko, Solid State Comm. 48, 675 (1983)
7. "Picosecond Modulated Reflectance in Semiconductors", A.V. Nurmikko, in Ultrafast Processes in Semiconductors, R.R. Alfano ed., vol.11, p. 509, Academic Press (1984)

8. "Kinetics of Free and Bound Excitons in Semiconductors", X.-C. Zhang, Y. Hefetz, and A.V. Nurmikko, in *Ultrafast Phenomena IV*, Springer Series in Chemical Physics, v. 38, p. 176 (1984)
9. "On the Question of Exciton Localization in (Cd,Mn)Te by Alloy disorder", X.-C. Zhang and A.V. Nurmikko", *Proc. 17 Int. Conf. Physics of Semiconductors*, North-Holland, p. 1443 (1985)
10. "Bound Exciton Formation and Excitonic Localization in Semimagnetic Semiconductors", A.V. Nurmikko, *J. of Luminescence* 30, 355 (1985)
11. "Picosecond Excitonic Phenomena in Semimagnetic Semiconductors", X.-C. Zhang, S.-K. Chang, and A.V. Nurmikko, *SPIE Proceedings* vol. 533, p.18 (1985)
12. "Photoluminescence in PbTe-PbEuTeSe Multiquantum Wells, W. Goltsos, J. Nakahara, A.V. Nurmikko, and D.L. Partin, *Appl. Phys. Lett.* 46, 1173 (1985)
13. "Electron-Hole Recombination Spectra and Kinetics in PbTe/PbEuTeSe Multiple Quantum Wells", W. Goltsos, J. Nakahara, A.V. Nurmikko, and D. Partin, *Proc. Int. Conf. Modulated Semiconductor Structures, Kyoto, Surface Science* (in press)
14. "Optical Bandgap and Magneto-Optical Effects in (Pb,Eu)Te", W.C. Goltsos, A.V. Nurmikko, and D.L. Partin, submitted to *Solid State Comm.*

In addition to these scientific publications, the AFOSR supported work has been presented in numerous scientific conferences (numbering in excess of twenty). Below is a sampling of invited presentations only.

- Conference on Progress in Quantum Electronics, Snowbird, Utah, Jan. 1982
- Conference on Mercury-Cadmium Telluride, Minneapolis, Feb. 1982
- General Motors Research Labs, Physics Department Colloquium, Detroit, March 1982
- Max Planck Institute for Solid State Physics, Colloquium, Stuttgart Germany, June 1982
- American Physical Society, Southeastern Section, Lexington, KY Oct. 1982
- IBM Research Laboratories, Physics Colloquium, Yorktown Heights NY, Nov. 1982
- Bell Laboratories, Seminar, Holmdel NJ, April 1983
- M.I.T. Magnet Laboratory, Seminar, Cambridge MA, May 1983
- Technical University of Helsinki, Colloquium, Finland, June 1983
- Optical Society of America Annual Meeting, New Orleans, October 1983
- 8th Int. Conf. of IR and MM-waves, Miami, December 1983

- University of Maryland, Physics Department Seminar, December 1983
- Northeastern University, Physics Colloquium, May 1984
- Purdue University, Electr. Engin. and Physics Seminar, May 1984
- Symposium on Semimagnetic Semiconductors, Bad Honnef, Germany, June 1984
- Conference on High Excitation and Ultrafast Phenomena in Semiconductors, Trieste, Italy, July 1984
- Topical Meeting on Semimagnetic Semiconductors, MIT, Cambridge, Oct. 1984
- Philips Research Laboratories, Colloquium, Briarcliff Manor NY, Nov. 1984
- Gordon Research Conference (International) on Nonlinear Optics, Brewster NH, July 1985
- International Symposium on Semimagnetic Semiconductors, Aussois France, September 1985

4. Personnel

The following have had direct support from this AFOSR grant:

- Prof. A.V. Nurmikko, Principal Investigator
- Dr. Michael Gal, a postdoctoral research associate in 1983, now at University of Sidney, Australia
- Bradley D. Schwartz, received Ph.D. degree 6/82; now at McDonnell Douglas Optoelectronics Center
- Carmen A. Huber, received Ph.D. degree 9/83; now with the University of Puerto Rico
- William C. Goltzos, is about to receive his Ph.D.

5. Patents

NO patents have been filed in connection with this AFOSR sponsored research

6. Remaining Funds

No remaining funds exist

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